# Optical properties of aerosols during APEX and ACE-Asia experiments

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[1] Sun/sky photometry and polarimetry of atmospheric light have been undertaken by multispectral photometers (CE-318-1 and -2, Cimel Electronique, France) and a polarimeter (PSR-1000, Opto Research, Japan) over Amami, Noto, and Shirahama, Japan, during APEX-E1, -E2, and ACE-Asia field campaigns. Radiometers provide us with the optical thickness of aerosols and Angström exponent. Other aerosol characteristics, e.g., size distribution, refractive index, etc., are retrieved based on each inversion method corresponding each equipment. The former takes a standard AERONET processing, and the latter is according to our own procedure to analyze the polarimetry with PSR-1000. After several aerosol parameters are derived, the HYSPLIT4 backward trajectory analysis is adopted to search the origin of aerosols. It is shown from these ground measurements that aerosol optical thickness, Angström exponent, and refractive index are classified into two typical categories as a background type detected in winter, and a soil dust type appeared in Asian dust events in spring. Further, it is found that the obtained size distribution of Asian dust indicates the dominance of large particles. INDEX TERMS: 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0394 Atmospheric Composition and Structure: Instruments and techniques; 1610 Global Change: Atmosphere (0315, 0325); 1640 Global Change: Remote sensing; KEYWORDS: aerosol optical thickness, Asian dust, AERONET, ACE-Asia, APEX

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## Introduction

- [2] Radiative parameters of atmospheric aerosols, such as optical thickness, size of particles, refractive index and albedo for single scattering, are well known to be important for radiation budget of Earth atmosphere-surface system. It is also known that Asia is the most complicated region for aerosol study, because emission of aerosols, e.g., sulfuric, nitric, carbonaceous, is increasing due to the economic growing. The soil dust, which is named Kosa, Yellow sands or Asian dusts, is also famous and increasing over Asia.
- [3] Asian Atmospheric Particle Environmental Change Studies (called APEX hereafter) experiment-1 and -2, and Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) intend to monitor the characteristic aerosols over East China Sea and Japan in early spring. It has been already pointed out that aerosols play a significant role as the cloud condensation nuclei (CCN) for low-level clouds [Nakajima et al., 2001]. The goal of these field experiments is better understanding for direct and indirect

2.1. Noto Site

will be shown first (see Table 1).

[5] Noto site (37.33°N, 137.13°E) is located at the center

of Noto peninsula in Japan. Noto peninsula is beaked into the Sea of Japan (refer to Figure 1), and there are no huge industries there. Therefore the aerosols seem to be typically oceanic in usual and occasionally contaminated with the continental compounds transported from Korea and China. The Cimel CE-318-2 Sun/sky photometer was operated there at 200m heights above sea level during APEX-E2/ ACE-Asia period as a temporary AERONET site.

effect of radiation forcing. In order to estimate such an effect,

the optical, chemical, microphysical properties of aerosols

are required. This work shows the aerosol properties over

the east Asia region based on Sun/sky photopolarimetric

**Overview of Ground-Based Measurements** 

[4] Three types of radiometers are operated during

APEX-E2 and ACE-Asia period. Each Sun photometer

was installed, respectively, at Noto, Shirahama and Amami

in Japan. Brief description of each site and each instrument

measurements in APEX and ACE-Asia periods.

# 2.2. Shirahama Site

[6] Shirahama site (33.68°N, 135.35°E) is facing the Pacific Ocean (refer to Figure 1). This site is about 200 km

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	Observational Sites		
Field campaigns	Noto, 37.33°N,	Shirahama, 33.68°N,	Amami-Oshima Island,
	137.13°E	135.35°E	28.37°N, 128.50°E
APEX-E1	n/a	CE-318-1	PSR-1000
APEX-E2/ACE-Asia	CE-318-2	CE-318-1	PSR-1000
Observing wavelength	0.44, 0.67, 0.87, 0.94,	0.34, 0.38, 0.44, 0.50, 0.67,	0.443, 0.49, 0.565, 0.67,
observing wavelength	and 1.02 μm, and polarization at 0.87 μm	0.87, 0.94, and 1.02 μm	0.765, and 0.865 μm with polarization at all bands

Table 1. Instruments and Observational Sites During APEX-E1/E2 and ACE-Asia

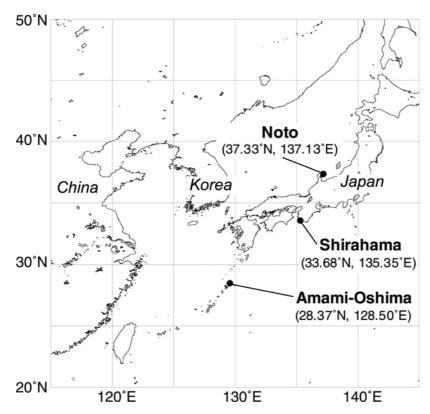
far away from huge cities as Kobe, Osaka and Kyoto, what one calls Kansai megalopolis. The Cimel CE-318-1 Sun/sky photometer has been worked at Shirahama since October 2000. Both Cimel radiometers of Noto and Shirahama were comparatively calibrated on site with standard AERONET instrument CE-318-1, which was freshly calibrated at NASA/GSFC, on 25 February 2001.

### 2.3. Amami-Oshima Island Site

[7] The site of Amami-Oshima Island (called Amami hereafter) is located at  $28.37^{\circ}N,\,128.50^{\circ}E$  in the East China Sea. The site is selected as the super site of APEX-E1/E2 projects. Various instruments had been set up during the period. A portable photopolarimeter PSR-1000 was operated to measure optical thickness of aerosols and the polarization information of diffused (sky) light. It has six observing wavelengths which are corresponding to ADEOS/POLDER sensor, i.e., 0.443, 0.490, 0.565, 0.670, 0.765 and 0.865  $\mu m$ . Polarimetric information are obtained with a Glan-Thompson polarizer. The Glan-Thompson polarizer manually rotates at

any direction away from the solar direction in the principal plane determined by the Sun and observer. In this work two angles were selected, i.e., 0-90 degrees system as for the linear polarization measurements. Then, degree of linear polarization is obtained by using maximum and minimum intensity of skylight. The observation with PSR-1000 involves two schemes as the direct sunlight measurement and the observation of polarized atmospheric light at six wavelength bands.

[8] For calibration of PSR-1000, Langley plot method is adopted [Shiobara et al., 1996; Sano et al., 2001]. Langley plot is a method to obtain the calibration constants of a radiometer based on the measurements of direct solar radiation on clear and stable days. The extraterrestrial values of PSR-1000 are obtained by using modified Langley method, which corrects the effect of gas absorption such as Ozone at 0.490, 0.565 and 0.670  $\mu$ m and strong O2-A-band absorption at 0.765  $\mu$ m. The transmittance by gaseous absorption is referred to AFGL model of LOWTRAN-7 [Kneizys et al., 1988]. The Langley plot's data of PSR-1000

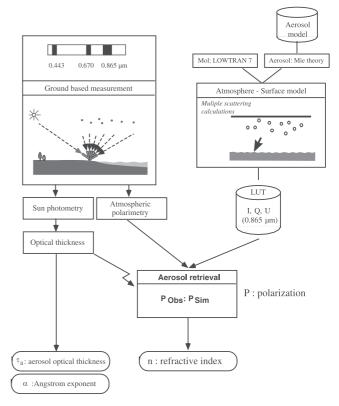


**Figure 1.** Geographical position of observational sites for APEX-E1, E2 and ACE-Asia. Noto and Shirahama sites are located in main island of Japan. Amami site is located at Amami-Oshima Island in the East China Sea.

for APEX-E1, E2 and ACE-Asia periods were obtained at Perth, Australia, in February 2000 at Mt. Haleakala in Hawaii in February 2001 and at Mt. Mauna Loa in Hawaii in March 2002.

### 3. Retrieval Algorithm for PSR-1000

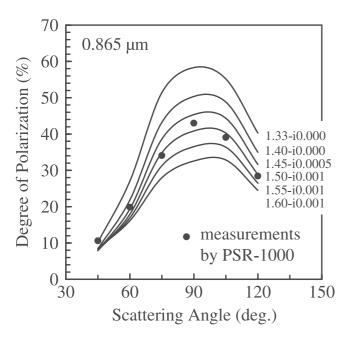
- [9] This section shows aerosol properties over Japan during APEX-E1/E2 and ACE-Asia period. In addition to aerosol optical thickness ( $\tau_a$ ) and Ångström exponent ( $\alpha$ ), the following parameters are also retrieved: (1) size distribution, refractive index, etc., for Noto; (2) size distribution, refractive index, etc., for Shirahama; and (3) refractive index for Amami.
- [10] The optical properties of Noto and Shirahama are derived from AERONET standard process [Holben et al., 1998; Dubovik et al., 2000; Dubovik and King, 2000]. The polarimetric measurements at Amami with PSR-1000 are analyzed according to our own procedure. Our retrieval algorithm is based on comparison of the observed data by PSR-1000 with the simulated results for multiple light scattering in the Earth atmosphere-ocean system. We tried to derive the refractive index (n) of aerosols from the polarization degree (%) at wavelength of 0.865 μm. Because polarization information is more sensitive to chemical composition of aerosols, particle size and irregularity of aerosol shape than radiance data [Mishchenko and Travis, 1997]. Moreover, degree of polarization takes an advantage to be a relative value. This means that degree of polarization does not need an absolute calibration of the equipment. That is why we use the degree of polarization for retrieval of refractive index (n). Further the reason why a wavelength of 0.865 µm is selected for the present retrieval of aerosol refractive index is based on the facts that radiance out of the ocean is very little, and optical thickness of atmospheric molecules is small there. At present work, oceanic model is assumed to be completely absorbent for clear water and Case II water reflectance is considered for the coastal region. These assumptions will be evaluated later.
- retrieval. Our system involves four blocks as; observation block, simulation block, retrieval block and reference data. Aerosol models in our simulations are described by the following parameters: (1) concentration, optical thickness of aerosols  $(\tau_a)$ ; (2) size, Ångström exponent  $(\alpha)$ , which is calculated from Mie-scattering theory assuming bimodal size distribution; and (3) chemical composition, complex refractive index (m). It is clear from Figure 2 that the values of  $\tau_a$  and  $\alpha$  are provided from the measurements of the direct sunlight. Thus our problem becomes to retrieve one parameter (n) in terms of polarization degree.
- 12] Dubovik et al. [2002a] have suggested such numerical values of parameters for size distribution as  $r_{\rm vf}=0.15~\mu m$ , and  $\sigma_f=0.42~\mu m$  for fine mode, and  $r_{\rm vc}=2.54~\mu m$  and  $\sigma_c=0.61~\mu m$  for coarse mode based on numerous AERONET measurements for desert dust and oceanic particles. Therefore our bimodal size distribution for aerosol models takes these values. S. Ohta (personal communication, 2002) measured the imaginary part of complex refractive index for Asian dust in the laboratory. The typical value of imaginary part is  $\sim\!0.001$  at wavelength of 0.865 μm. This value of  $\sim\!0.001$  for the imaginary part



**Figure 2.** System flow for retrieval of refractive index from Sun radiometry and sky polarimetry for PSR-1000 measurements at Amami-Oshima Island.

coincides with the value suggested by *Dubovik et al.* [2002a] derived from AERONET observations. Thus numerical values of complex refractive index of aerosols for simulations are set up ranging from 1.33-i0.000 to 1.60-i0.001. Including the above aerosol models, Earth atmosphere-sea surface-ocean model is set up because of Amami site locates near the coast. The sea surface is simulated by multiple facets with slopes that vary according to an isotropic Gaussian distribution with respect to wind speed, as described by Cox and Munk [1954]. We found from numerical simulations that changing of wind speed from 3 m/s to 7 m/s provides the influence of less than  $\pm 0.1\%$  upon the retrieved refractive index. In this study, the typical value for wind speed on a clear day is assumed to be 7 m/s. Not only the contribution to the water leaving radiance from the sea surface reflectance but also that from the ocean itself should be considered. The turbid water case is examined using the coastal data of the reflectance value of  $\sim 0.62\%$  at a wavelength of  $0.865 \,\mu m$  [Liew et al., 2001]. It is found that the discrepancy between the turbid water case and the complete absorbent case is very little, i.e., the effect is less than  $\pm 0.1\%$  in refractive index at a wavelength of 0.865 µm. Molecular information such as gaseous transmittance and absorption is determined by referring to AFGL U.S. standard from LOWTRAN 7 [Kneizys et al.,

[13] The radiative transfer equation is calculated by the doubling-adding method [*Mukai et al.*, 1992; *Sano and Mukai*, 1998]. Computations are performed for 25 zenith Sun and PSR (viewing) angles and 0-Pi azimuth angles.



**Figure 3.** Relationship of polarization degree versus scattering angle at a wavelength of  $0.865 \, \mu m$  where aerosol optical thickness and Angstrom exponent are fixed.

Figure 3 shows the relationship between degree of polarization at a wavelength of 0.865  $\mu m$  and scattering angle. The solid curves represent the simulated values for various refractive indices, where optical thickness and Ångström exponent are obtained from Sun photometry. The filled circles show the example of polarization degree measurements with various scattering angles from 45 degree to 120 degree. From this figure, we can estimate the refractive index from the measurements of Sun photometry and at least a polarization degree measurement. In this work, the measurements which are undertaken at the wavelength of 0.865  $\mu m$  and scattering angles of 75 or 90 degree is selected because of small water leaving radiance from the ocean and maximum sensitivity for refractive indices.

# 4. Aerosol Properties at Noto, Shirahama, and Amami

### 4.1. APEX-E1 Experiment in December 2000

[14] The APEX Experiment 1 (E1) was done from 5–23 December 2000. The measurements of aerosol particles at Amami are shown in Figures 4a and 4b, which present optical thickness of aerosols and Ångström exponent ( $\alpha$ ) which is calculated from the spectral tendency of optical thickness of aerosols as below:

$$\alpha = -\ln(\tau_{\lambda_1}/\tau_{\lambda_2})/\ln(\lambda_1/\lambda_2), \tag{1}$$

where wavelengths  $\lambda_1$  and  $\lambda_2$  take values of 0.443 and 0.865, or 0.870  $\mu m$ , respectively. The values of  $\alpha$  are closely related to the aerosol size distribution. For example, the small values of  $\alpha$  indicate the large particles such as oceanic aerosols and dust aerosols. On the contrary, the large values represent small particles such as artificial aerosols. In general, the values of Ångström exponent ( $\alpha$ ) from  $\sim$ 0 to 1 shows coarse particles (such as sea salt solution, and soil

dusts), on the contrary,  $1 < \alpha < \sim 2.5$  indicates small particles (such as sulfate, biomass burning etc.). The gap in Figure 4 represents the lack of Sun photometry due to the weather conditions such as cloudy and/or rainy. The aerosol optical thickness shows typical values of Amami region, i.e., averaged optical thickness at wavelength at 0.443, 0.670, and 0.865  $\mu$ m are the 0.17, 0.10, and 0.09, respectively. The averaged Angström exponent is 1.02 which indicates that size of aerosols are mixed with both small and coarse particles at Amami in December 2000.

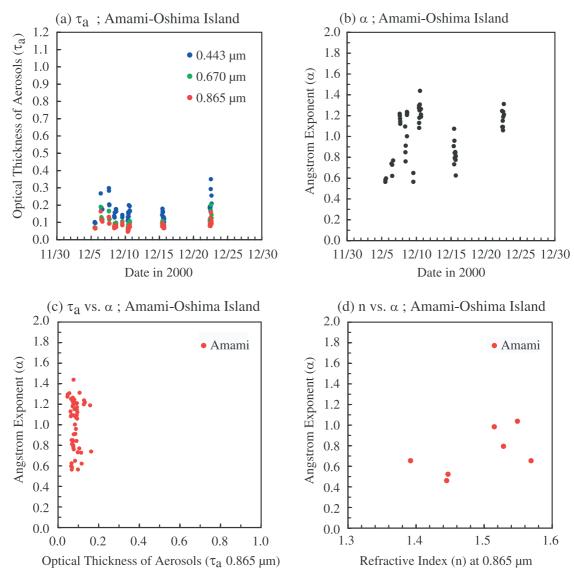
[15] Figures 4c shows a scattergram between Ångström exponent ( $\alpha$ ) and aerosol optical thickness ( $\tau_a$ ) at a wavelength of 0.865  $\mu m$  in December 2000. Typical background aerosol characteristics over Amami region are shown here. Namely, the optical thickness is not so changeable but the variance of Ångström exponent is large. Figure 4d shows the refractive index retrieved according to the procedure of Figure 2, where optical thickness of aerosols at a wavelength of 0.865  $\mu m$ , Ångström exponent, and polarization degree at 0.865  $\mu m$  are used.

[16] Figure 4d is roughly divided into two groups as  $n \sim 1.4$  and  $n \sim 1.5$ . It seems that the first group corresponds to the transparent particles as oceanic or sulfuric aerosols, and the second one represents the polluted one. These results suggest the typical aerosol condition over Japan in winter time. Figure 5 presents spectral polarization degree for reference. Each symbol in Figure 5 shows the different aerosol conditions, i.e., open squares represent the polarization degree of background aerosol condition in winter. The filled circles denote the measurements undertaken on dust free day in spring. The open circles correspond to the Asian dust on 14 April, which will be interpreted in detail in next section. From Figure 5, we found that heavy aerosol loading causes decreasing of the polarization degree due to the multiple scattering effects.

### 4.2. APEX-E2/ACE-Asia Experiments in April 2001

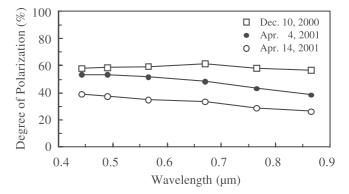
[17] The APEX Experiment 2 (E2) and ACE-Asia field campaigns were done in April 2001. Two Cimel Sun/sky radiometers and a polarimeter (PSR-1000) were operated at three different places (refer to Figure 1) during the experiments. The Sun photometric results are shown in Figure 6, where the aerosol optical thickness is in Figures 6a, 6b, and 6c, and its Ångström exponent is in Figures 6a', 6b', and 6c' in April 2001. The trend of optical thickness of aerosols shows the typical monsoon pattern in east Asia, that is, the values in winter is much smaller than those in spring (refer to Figure 4). The averaged values of  $\tau_a$  at a wavelength of 0.870  $\mu m$  over Asian dust period alone of Figure 6 are 0.37, 0.33, and 0.34, respectively, at Noto, Shirahama, and Amami. The same values of  $\tau_a$  but for usual time are 0.15, 0.12, and 0.12. It is found that aerosol optical thickness in Asian dust event is more than double of usual value. This result coincides with the satellite product [Sano and Mukai, 2000].

[18] Now individual Asian dust event is considered. In fact, high values of aerosol optical thickness appear on many days in Figure 6. We know Asian dust events frequently occurred in spring, 2001. For example, the dust events are observed on 3, 6, 11, 24, and 26 March and on 10 and 14 April at Shirahama site. Some of them are also detected at Noto and Amami sites, but no other events simultaneously occur with each other. Although the Ångström exponent in

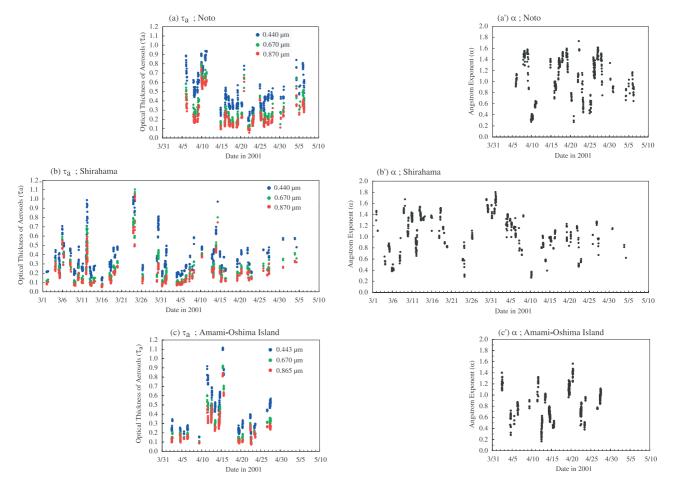


**Figure 4.** (a) Optical thickness of aerosols  $\tau a$  at wavelengths of 0.443, 0.670, and 0.865  $\mu m$ . (b) Angstrom exponent  $\alpha$ . (c) Relationship between  $\alpha$  and  $\tau a$  at wavelengths of 0.865  $\mu m$ . (d) Relationship between refractive index (n) and  $\alpha$ . The measurements were undertaken at Amami-Oshima Island (28.37°N, 128.50°E) by PSR-1000 during APEX-E1 in December 2000.

spring is totally variable, the averaged value looks a good indicator for dust event. Ångström exponent takes small values in dust event. Namely Angström exponent averaged over the dust event alone takes the value of 0.76, 0.80, and 0.66 at Noto, Shirahama, and Amami, respectively. On the other hand, the value averaged over the dust free days takes 1.3, 1.2, and 0.95 at Noto, Shirahama, and Amami, respectively. Similar feature is also detected in a scattergram between aerosol optical thickness versus Angström exponent in Figure 7a. Each color, i.e., green, blue, red, indicates the measurements of each site. We found in Figure 7a that the value of Angström exponent is decreasing with optical thickness, especially in large optical thickness region, where several characteristic clustering classes are found. Each class seems to represent each dust event. That is, small values of Angström exponent represent dust particles and large ones correspond to haze particles. This subject will be discussed later.



**Figure 5.** Degree of linear polarization at a scattering angle of 90 degrees away from the Sun in such typical three days as a winter day (10 December 2000), an ordinal spring day (4 April 2001) and Asian dust day (14 April 2001) at Amami-Oshima Island (28.37°N, 128.50°E) by PSR-1000.



**Figure 6.** (a, b, and c) Optical thickness of aerosols at wavelengths of 0.443, 0.670, and 0.865 μm and (a', b', and c')Angstrom exponent. The measurements were undertaken at Noto (37.33°N, 137.13°E) by Cimel CE-318-2 Sun/sky radiometer, Shirahama site (33.68°N, 135.35°E) by Cimel CE-318-1, Amami-Oshima Island (28.37°N, 128.50°E) by PSR-1000 during APEX-E2/ACE-Asia period from March to April 2001.

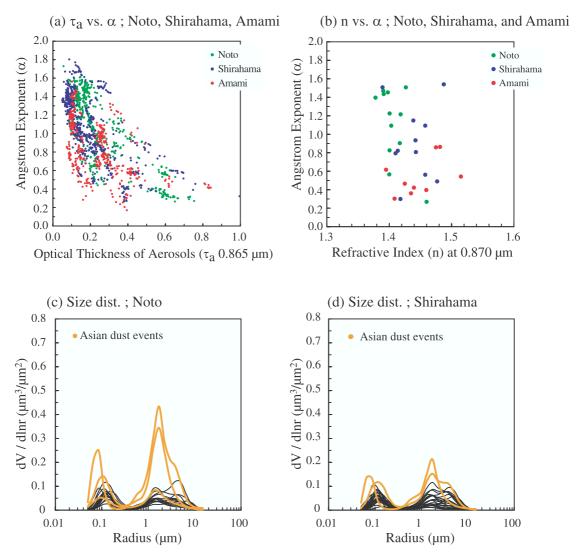
[19] The retrieved refractive index of aerosols is discussed now. Note that the values in Figure 7b represent the daily averaged values, and refractive index denotes the value at a wavelength of 0.870 µm at Amami, Noto, and Shirahama, respectively. The reason why we choose a wavelength of 0.870 µm that affection of nonsphericity by dust particles is small artifacts rather than that of shorter wavelengths and are correct for desert dust even if spheres are assumed in the retrieval [Dubovik et al., 2002b]. It is found that refractive index over Japan changes from 1.4 to 1.5, and the representative value is 1.43. This fact suggests that the dust particles over Japan are mixed with various components, i.e., oceanic particles, anthropogenic particles from the continent [Takemura et al., 2000]. The satellite results also indicate the existence of the small anthropogenic particles over southeast Asia in spring [Deuzé et al., 2001; Sano et al., 2002]. Figures 7c and 7d represent the retrieved results of volume size spectrum. The brown and black curves represent the results during dust events and dust free period, respectively. We found that the radius of dust particles exists between 1 µm and 2 µm. Namely coarse particles are dominant during the dust events. Note that concentration of very small particles (radius  $< 0.1 \mu m$ )

are overestimated during dust events due to nonsphericity artifacts. These artifacts will be removed with using retrieval based on spheroids [Dubovik et al., 2002b].

### 4.3. Transportation of Aerosol Particles

[20] As shown above, aerosol parameters (e.g., optical thickness of aerosols and Ångström exponent) are retrieved from ground-based observation. Together with these two aerosol parameters retrieved on the ground, information on the transportation of aerosol particles can be utilized to better understand the characteristics of aerosol particles. HYSPLIT4 (Hybrid Single-Particle Lagrangian Integrated Trajectory) Model developed by NOAA/Air Resources Laboratory (see http://www.arl.noaa.gov/ready/hysplit4. html) can be utilized to analyze the particle transportation. Meteorological data such as air pressure, u and v component of wind, geopotential height, etc, are needed as input data for the calculation with HYSPLIT4 model. NCEP/NCAR Reanalysis data with 2.5-degree grid and with the resolution of 6-hours interval are used as input meteorological data into HYSPLIT4 model in this study.

[21] The calculations of particle transportation are conducted for three sites (i.e., Noto, Shirahama and Amami).



**Figure 7.** (a) Relationship between Angstrom exponent ( $\alpha$ ) and optical thickness of aerosols ( $\tau_a$ ) at a wavelength 0.865  $\mu$ m. (b) Refractive index (n) at a wavelength of 0.870  $\mu$ m versus  $\alpha$ . The green, blue, and red color represent Noto, Shirahama, and Amami measurements during APEX-E2/ACE-Asia in April 2001. (c) Volume size distribution at Noto site during APEX-E2/ACE-Asia. Brown curves represent the volume distribution during Asian dust events. (d) Same as Figure 7c but at Shirahama.

Hereafter, some interesting features of aerosols are introduced combining the results of ground-based observation and that from HYSPLIT4 calculation (refer to Figure 8).

[22] Analysis of the data observed at Noto site on 10 and 11 April 2001 show similar high values in optical thickness of aerosols with a little difference in Ångström exponent (see Figure 6). Only with these two parameters of aerosols, types of aerosols are difficult to be distinguished. However, results of HYSPLIT4 backward trajectories in Figures 8a and 8b show totally different transportation of the air masses between these two days. That is, on 10 April in Figure 8a, the air masses mainly come from the China continent passing through east Asia, and hence include large sized aerosols generated around eastern Asia. On 11 April in Figure 8b, on the other hand, aerosol particles observed around Noto site mainly come from the eastern direction via the Pacific Ocean, where lots of oceanic aerosols may be produced. The reason of high optical thickness of aerosols

is considered as the long distance transportation and/or sulfur dioxide (SO<sub>2</sub>) passing over Miyake-jima volcano [*Ohta et al.*, 2001].

[23] Around Shirahama various days with high optical thickness of aerosols are seen on March and April 2001 (refer to Figure 6). Among them, the optical thickness of aerosols on 24 and 27 March takes much higher values. The value of optical thickness of aerosols at 0.865 μm exceeds 0.5. From the results of backward trajectories by HYSPLIT4 model in Figures 8c and 8d, air masses are coming from the China continent passing around Gobi desert. In this case, combined analysis of aerosol parameters from ground-based measurement and trajectories analysis has clearly revealed the desert dust reached to and observed in the station of Shirahama.

[24] The observations at Amami Island from the beginning to the end of April show the period of high optical thickness of aerosols from 11 to 15 April (in Figure 6).

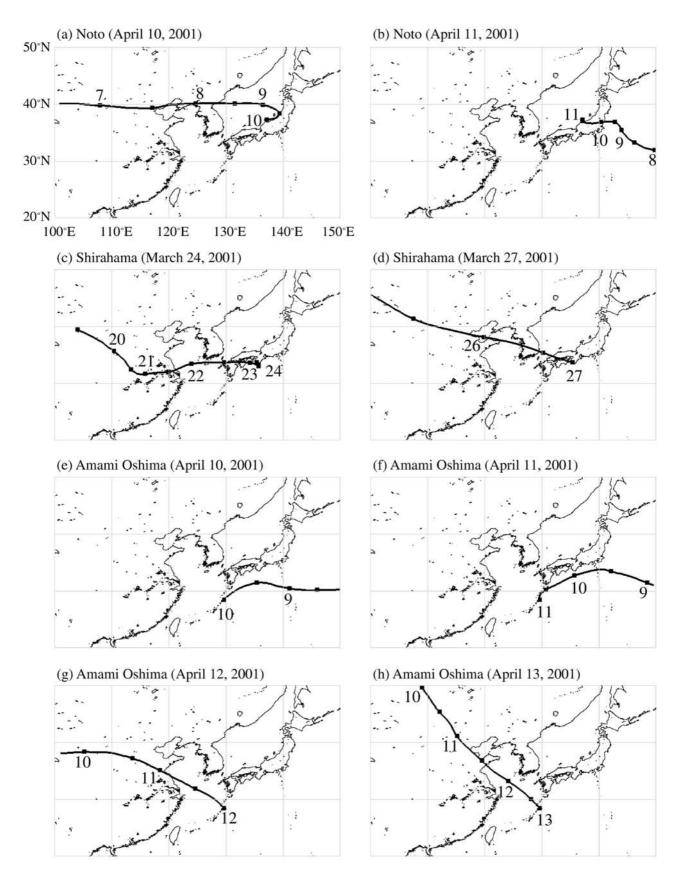


Figure 8. Transportation map of particles calculated with HYSPLIT4 model for (a and b) Noto, (c and d) Shirahama, and (e, f, g, and h) Amami.

Figures 8e–8h correspond to the results of backward trajectories by HYSPLIT4 for 10–13 April, respectively. It is clearly seen that the aerosol optical thickness is different from background one during this period (e.g., compared with the data of days other than 10–15 April). However, even the similar high values in optical thickness turned out to have different origin of the aerosol particles. The results calculated with HYSPLIT4 show the transportation of particles from eastern direction for 10 and 11 April, and those from the China continent are seen from 12 and 13 April, which indicates the very high temporal variability of aerosol particles but with similar optical thickness during the short period.

[25] Even the results for the similar westward transportation on 10 and 11 April show different northerly and southerly westward wind. Although the observation data is not available for 10 April, the pathway of this day shows the possibility of the different contamination of sulfur dioxide (SO<sub>2</sub>) passing over the volcano Miyake-jima for the two days.

### 5. Conclusions

- [26] The aerosol optical thickness and Ångström exponent in dust season have been measured. The optical properties of aerosols, such as refractive index (all sites) and size distribution (at Noto and Shirahama) are retrieved from light scattering simulations. It is found that aerosol optical thickness in Asian dust event is more than double of usual value, and Ångström exponent takes small values in dust event. This result coincides with the satellite product.
- [27] Our retrieval method based on the polarimetry will be improved for explaining dust particles with nonsphericity effect [*Dubovik et al.*, 2002b] in the further work. At any rate, we found that polarization data is available for estimation of refractive index of aerosols.
- [28] Spectral Radiation-Transport Model for Aerosol Species (SPRINTARS) [Takemura et al., 2000] simulates the production, transport of aerosols with atmospheric general circulation model (AGCM). Takemura et al. [2002] have pointed out that large amount of sulfate are dominant at Noto from 7-9 and 11-19 April 2001. Half of optical thickness are occupied by dust particles and Ångström exponent takes the value of  $\sim 0.3$  on 10 April in spite of large values in usual. This interpretation coincides with our ground measurements.
- [29] Information on aerosol transportation using HYSPLIT4 model is used together with the analyzed data of ground-based observation. The results of HYSPLIT4 and aerosol optical parameters show similar optical thickness of aerosols but with different origin of the particles for different days.
- [30] Acknowledgments. The authors wish to greatly acknowledge T. Nakajima, University of Tokyo, for contribution to take the measurements at Amami under the JST-CREST/APEX (Asian Atmospheric Particle Environmental Change Studies) project. The AERONET group (R. Nelson, W. Newcomb, A. Scully, A. Vermeulen, O. Dubovik, and I. Slutsker) also kindly supported the calibrations and processing the measurements both of Noto and Shirahama. The authors wish to thank to M. Yasumoto, T. Kitajima, Kinki University, and H. Muroishi of Yanagida Observatory for their contributions to take/maintain measurements at Shirahama, Amami-Oshima, and Noto sites. Additional thanks to two reviewers for valuable comments on this study. HYSPLIT4 (Hybrid Single-Particle Lagrangian Integrated Trajectory) Model was provided from http://www.arl.noaa.gov/ready/hysplit4.html, NOAA Air Resources Laboratory. This work was supported in part by grants-in-aid for Young Scientists (A) (14702069; 2002) and Scientific Research on Priority Areas (14048201; 2002) from the Ministry of Education, Culture, Sports, Science and

Technology (MEXT), and grant for Environmental Research Projects (023069; 2002) from the Sumitomo Foundation.

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